

February 28, 2002

## BOX PCT

Commissioner for Patents  
Washington, D.C. 20231

PCT/JP00/05780  
-filed August 28, 2000

Re: Application of Etsuo MINAMINO, Kazuyoshi MIMURA, and  
Mitsuhiro OTANI  
ULTRAVIOLET-CROSSLINKABLE POLYMER COMPOSITION  
**Assignee: DAIKIN INDUSTRIES, LTD.**  
Our Ref: Q68612

Dear Sir:

The following documents and fees are submitted herewith in connection with the above application for the purpose of entering the National stage under 35 U.S.C. § 371 and in accordance with Chapter II of the Patent Cooperation Treaty:

- ☒ an executed Declaration and Power of Attorney.
- ☒ an English translation of the International Application.
- ☒ Notification Concerning Submission or Transmittal or Priority Document.
- ☒ an executed Assignment and PTO 1595 form.
- ☒ International Search Report and references cited in the ISR.
- ☒ an Information Disclosure Statement and PTO 1449 form with references.
- ☒ a Preliminary Amendment

It is assumed that copies of the International Application, the International Search Report, the International Preliminary Examination Report, and any Articles 19 and 34 amendments as required by § 371(c) will be supplied directly by the International Bureau, but if further copies are needed, the undersigned can easily provide them upon request.

The Government filing fee, after entry of the Preliminary Amendment, is calculated as follows:

|                                  |           |             |                 |
|----------------------------------|-----------|-------------|-----------------|
| Total claims                     | 11 - 20 = | x \$18.00 = | \$0.00          |
| Independent claims               | 1 - 3 =   | x \$84.00 = | \$0.00          |
| Base Fee                         |           |             | \$890.00        |
| <b>TOTAL FILING FEE</b>          |           |             | <b>\$890.00</b> |
| <b>Recordation of Assignment</b> |           |             | <b>\$ 40.00</b> |
| <b>TOTAL FEE</b>                 |           |             | <b>\$930.00</b> |

Checks for the statutory filing fee of \$890.00 and Assignment recordation fee of \$40.00 are attached. You are also directed and authorized to charge or credit any difference or



SUGHRUE MION, PLLC

National Stage of PCT/JP00/05780  
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overpayment to Deposit Account No. 19-4880. The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16, 1.17 and 1.492 which may be required during the entire pendency of the application to Deposit Account No. 19-4880. A duplicate copy of this transmittal letter is attached.

Priority is claimed from:

| <u>Country</u> | <u>Application No</u> | <u>Filing Date</u> |
|----------------|-----------------------|--------------------|
| Japan          | 244653/1999           | August 31, 1999    |

Respectfully submitted,

A handwritten signature in cursive script, appearing to read 'A. Rosner', positioned above a horizontal line.

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2003 FEB 28 10 06 AM '02

PATENT APPLICATION  
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Etsuo MINAMINO, et al.

Appln. No.: National Stage of PCT/JP00/05780

Confirmation No.: NOT YET ASSIGNED

Group Art Unit: NOT YET ASSIGNED

Filed: February 28, 2002

Examiner: NOT YET ASSIGNED

For: ULTRAVIOLET-CROSSLINKABLE POLYMER COMPOSITION

**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-identified application as follows:

**IN THE CLAIMS:**

Please enter the following amended claims:

4. (Amended) A crosslinked article obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 1 to ultraviolet irradiation.

5. (Amended) A gasket for magnetic disc production apparatus which is obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 1 to ultraviolet irradiation.

6. (Amended) A fluorine-containing polymer coating material comprising the ultraviolet-crosslinkable polymer composition of Claim 2.

PRELIMINARY AMENDMENT  
National Stage of PCT/JP00/05780

**Please add the following new claims:**

7. (New) A crosslinked article obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 2 to ultraviolet irradiation.
8. (New) A crosslinked article obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 3 to ultraviolet irradiation.
9. (New) A gasket for magnetic disc production apparatus which is obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 2 to ultraviolet irradiation.
10. (New) A gasket for magnetic disc production apparatus which is obtained by subjecting the ultraviolet-crosslinkable polymer composition of Claim 3 to ultraviolet irradiation.
11. (New) A fluorine-containing polymer coating material comprising the ultraviolet-crosslinkable polymer composition of Claim 3.

PRELIMINARY AMENDMENT  
National Stage of PCT/JP00/05780

REMARKS

Entry and consideration of this Amendment is respectfully requested.

Respectfully submitted,



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**APPENDIX**

**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

**IN THE CLAIMS:**

**The claims are amended as follows:**

4. (Amended) A crosslinked article obtained by subjecting the ultraviolet-crosslinkable polymer composition of ~~any of~~ Claims 1 ~~to~~ 3 to ultraviolet irradiation.
5. (Amended) A gasket for magnetic disc production apparatus which is obtained by subjecting the ultraviolet-crosslinkable polymer composition of ~~any of~~ Claims 1 ~~to~~ 3 to ultraviolet irradiation.
6. (Amended) A fluorine-containing polymer coating material comprising the ultraviolet-crosslinkable polymer composition of Claim 2 ~~or~~ 3.

**Claims 7-11 are added as new claims.**

## DESCRIPTION

## ULTRAVIOLET-CROSSLINKABLE POLYMER COMPOSITION

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## TECHNICAL FIELD

The present invention relates to a polymer composition having iodine and/or bromine which is crosslinkable by irradiation with ultraviolet rays.

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## BACKGROUND ART

15

For crosslinking a fluorine-containing polymer, particularly a fluorine-containing elastomer, usual crosslinking methods are polyamine vulcanization, polyol vulcanization and organic peroxide vulcanization. In any of those methods, a high temperature is necessary in order to obtain a crosslinked article and therefore an application thereof is limited.

20

Particularly in case of a method of crosslinking a fluorine-containing elastomer having iodine or bromine, generally a method of crosslinking with an organic peroxide is employed. However the crosslinking needs to be carried out at high temperature and in case of usual molding, a crosslinking reaction does not proceed in the neighborhood of an interface with oxygen and staining of a metal die arises. Thus there is a problem with processability.

25

JP-B-5-63482, etc. describe crosslinking methods other than the method of crosslinking by using heat energy, namely methods of crosslinking by using radiation, etc. Such methods require expensive facilities and are not carried out easily and therefore application thereof

is limited to only particular fields.

For example, in crosslinking of a fluorine-containing polymer, ionizing radiation, etc. have been adopted practically for crosslinking of a tetrafluoroethylene-ethylene copolymer (ETFE), polyvinylidene fluoride (PVdF), and the like, but only in the field of electric cables. As for perfluoropolymers, it is assumed that crosslinking thereof is still difficult.

On the other hand, urethane rubber and silicone rubber are on the market and are widely used because crosslinking need not be carried out at high temperature and can be carried out at room temperature. However the urethane rubber has a problem with heat resistance and water resistance, and the silicone rubber has a problem with chemical resistance and oil resistance.

As mentioned above, there have been no fluorine-containing polymer compositions which could be cured at room temperature while maintaining excellent properties of the fluorine-containing polymer.

An object of the present invention is to provide a crosslinkable composition comprising a polymer containing iodine and/or bromine in a specific amount, particularly an ultraviolet-crosslinkable fluorine-containing polymer, and also to provide a crosslinked article obtained from the composition.

#### DISCLOSURE OF INVENTION

The present invention relates to an ultraviolet-crosslinkable polymer composition which comprises a polymer containing 0.001 to 10 % by weight of iodine and/or bromine, a photoinitiator as a crosslinking agent and a polyfunctional unsaturated compound as a



crosslinking aid.

When the polymer is a fluorine-containing polymer, particularly a fluorine-containing elastomer, an excellent effect is exhibited.

5 Hitherto in crosslinking with ultraviolet rays, usually an acryl group is activated by irradiation of ultraviolet rays to become a crosslinking point. However in the present invention, iodine or bromine contained in the polymer is a crosslinking point. Such a crosslinking system has not been known and is a novel one.

#### BEST MODE FOR CARRYING OUT THE INVENTION

In the present invention the polymer having iodine and/or bromine may be a resin or an elastomer.

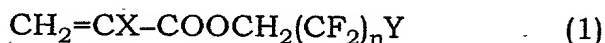
For introducing iodine or bromine into the polymer, various  
15 known methods can be used. For example, there are (1) a method of copolymerizing a monomer having iodine and/or bromine (hereinafter referred to as "Introducing Method 1". JP-A-8-53595, JP-B-5-63482, JP-A-4-288305, JP-B-53-4115, etc.); (2) a method of using an iodine or bromine compound as a polymerization initiator or a chain transfer  
20 agent (hereinafter referred to as "Introducing Method 2". JP-A-63-23907, JP-B-6-11773, JP-B-1-16844, JP-B-63-41928, JP-B-5-406, JP-A-62-36407, etc.), and the like. The introducing method is not limited to them.

First, a resin as a basic component is explained below.  
25 Either of a non-fluorine-containing resin and a fluorine-containing resin may be used as a resin.

Examples of the non-fluorine-containing resin are, for

instance, homopolymers or copolymers of poly(meth)acrylic acid esters such as polymethyl methacrylate and polyethyl acrylate. Examples of the monomer to be used are methyl acrylate, ethyl acrylate, methyl methacrylate, ethyl methacrylate, and the like. Examples of the comonomer are, for instance, ethylene, propylene, vinyl chloride, vinylidene chloride, acrylonitrile, vinyl esters such as vinyl acetate, vinyl ethers such as ethyl vinyl ether, vinyl monomers such as styrene, and the like.

Examples of the fluorine-containing resin are homopolymers or copolymers of fluorine-containing (meth)acrylic acid esters represented by the formula (1):



wherein X is H, F or  $\text{CH}_3$ , n is an integer of from 1 to 8, Y is H or F. Examples of the comonomer are the above-mentioned (meth)acrylic acid esters and/or vinyl monomers.

Also there are polytetrafluoroethylene, polyvinylidene fluoride, ethylene-tetrafluoroethylene copolymer, ethylene-chlorotrifluoroethylene copolymer, tetrafluoroethylene-hexafluoropropylene copolymer, tetrafluoroethylene-perfluoro(alkyl vinyl ether) copolymer, and the like.

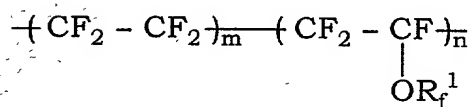
While the elastomer to be used in the present invention may be a non-fluorine-containing elastomer, particularly a fluorine-containing elastomer which has been unable to be crosslinked with ultraviolet rays can be used preferably. Examples of the fluorine-containing elastomer are, for instance, vinylidene fluoride copolymers

such as vinylidene fluoride-hexafluoropropylene copolymer, vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymer and vinylidene fluoride-chlorotrifluoroethylene copolymer; tetrafluoroethylene-propylene copolymer and ethylene-hexafluoropropylene copolymer; fluoro(alkyl vinyl ether) (encompassing those having plural ether bonds)-olefin copolymers such as vinylidene fluoride-tetrafluoroethylene-perfluoro(alkyl vinyl ether) copolymer and tetrafluoroethylene-perfluoro(alkyl vinyl ether) copolymer; alkyl vinyl ether-chlorotrifluoroethylene copolymer, and the like.

Also other monomer component copolymerizable thereto may be copolymerized. Examples thereof are vinyl chloride, vinylidene chloride, acrylonitrile, vinyl esters such as vinyl acetate, vinyl ethers such as ethyl vinyl ether, styrene, and the like.

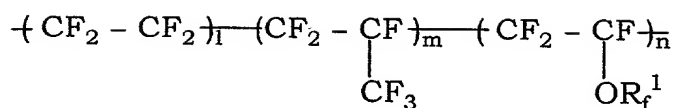
In the present invention the fluorine-containing elastomer may be the above-mentioned fluorine-containing elastomers and in addition, those having, as an elastomer chain, a perfluoro elastomer chain represented by the following formula (2) or (3) or a non-perfluoro elastomer chain represented by the formula (4), (5), (6) or (7).

A copolymer elastomer chain represented by the formula (2):



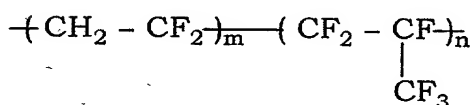
wherein m is from 95 to 50, n is from 5 to 50,  $R_f^1$  is a perfluoroalkyl group having 1 to 8 carbon atoms,

A terpolymer elastomer chain represented by the formula (3):



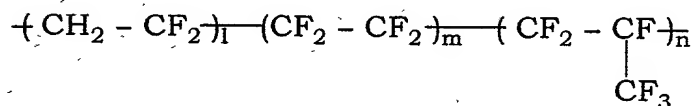
wherein l is from 95 to 35, m is from 0 to 30, n is from 5 to 35,  $R_f^1$  is a perfluoroalkyl group having 1 to 8 carbon atoms,

A copolymer elastomer chain represented by the formula (4):



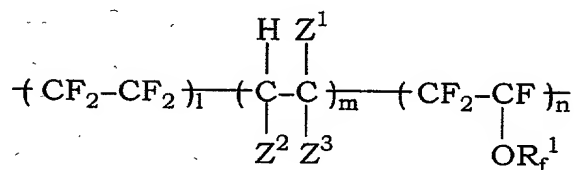
wherein m is from 85 to 60, n is from 15 to 40,

A terpolymer elastomer chain represented by the formula (5):



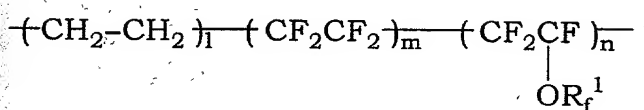
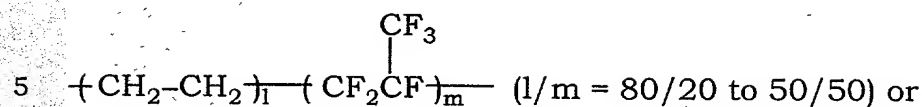
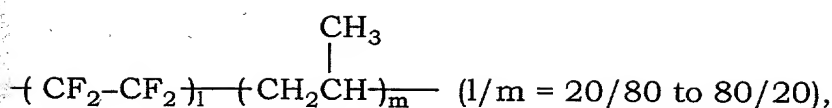
wherein l is from 85 to 20, m is from 0 to 40, n is from 15 to 40,

A terpolymer elastomer chain represented by the formula (6):



wherein l is from 95 to 45, m is from 0 to 10, n is from 5 to 45,  $Z^1$ ,  $Z^2$  and  $Z^3$  are fluorine atom or hydrogen atom independently,  $R_f^1$  is a perfluoroalkyl group having 1 to 8 carbon atoms, and

A non-perfluoro elastomer chain represented by the formula (7):



10 (l is from 1 to 80, m is from 0 to 80, n is from 10 to 50,  $R_f^1$  is as defined above).

Further there may be used a fluorine-containing multi-segment polymer prepared by block-copolymerizing the above-mentioned elastomer as an elastomer segment with a fluorine-  
15 containing resin as a hard segment.

Then the monomer for introducing iodine and/or bromine is explained below.

Examples of the monomer having iodine and/or bromine which can be used in Introducing Method 1 are monomers described in  
20 the above-mentioned JP-A-8-53595. Examples thereof are, for instance,  $\text{CH}_2=\text{CHCH}_2\text{OCOCF}_2\text{CH}_2\text{I}$ ,  $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2(\text{CF}_2)_4\text{I}$ ,  
 $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2\text{I}$ ,  $\text{CH}_2=\text{CHCH}_2\text{OCH}_2\text{CH}_2(\text{CF}_2)_4\text{I}$ ,  
 $\text{CH}_2=\text{CHCH}_2\text{OCH}_2\text{CH}_2\text{I}$ ,  $\text{CH}_2=\text{CHCH}_2\text{OCOCF}_2\text{CH}_2\text{Br}$ ,  
 $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2(\text{CF}_2)_4\text{Br}$ ,  $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2\text{Br}$ ,  
25  $\text{CH}_2=\text{CHCH}_2\text{OCH}_2\text{CH}_2(\text{CF}_2)_4\text{Br}$ ,  $\text{CH}_2=\text{CHCH}_2\text{OCH}_2\text{CH}_2\text{Br}$ , and the like.

It is preferable that those iodine- and/or bromine-containing monomers are used for the non-fluorine-containing polymers from the viewpoint of

copolymerizability.

Also there are monomers described in the above-mentioned JP-B-5-63482, JP-A-4-288305, JP-B-53-4115, etc. Examples thereof are, for instance,  $\text{CF}_2=\text{CFOCF}_2\text{CF}_2\text{CH}_2\text{I}$ ,  $\text{CF}_2=\text{CFOCF}_2(\text{CF}_3)\text{CFOCF}_2\text{CF}_2\text{CH}_2\text{I}$ ,  $\text{CF}_2=\text{CFI}$ ,  $\text{CF}_2=\text{CHBr}$ ,  $\text{CH}_2=\text{CHCF}_2\text{CF}_2\text{Br}$ ,  $\text{CF}_2=\text{CFOCF}(\text{CF}_3)\text{CF}_2\text{OCF}_2\text{Br}$ , and the like. It is preferable that those iodine- and/or bromine-containing monomers are used for the fluorine-containing polymers from the viewpoint of copolymerizability.

Polymerization methods which can be used for this introducing method are known methods such as emulsion polymerization, suspension polymerization, solution polymerization, and the like.

The monomer is copolymerized in such an amount that iodine and/or bromine are contained in the obtained polymer in an amount of from 0.001 to 10 % by weight, preferably from 0.05 to 7 % by weight.

Introducing Method 2 is a method of introducing iodine and/or bromine into an end of the polymer by using iodine and/or bromine or a compound having iodine and/or bromine as a polymerization initiator or a chain transfer agent.

Examples of such a polymerization initiator are, for instance, those described in the above-mentioned JP-A-63-23907. Concretely there are, for example,  $(\text{ICF}_2\text{COO})_2$ , and the like.

Examples of such a chain transfer agent are, for instance, those described in the above-mentioned JP-B-6-11773, JP-B-1-16844, JP-B-63-41928, JP-B-5-406, JP-A-62-36407, etc. Concretely there are,



for example,  $\text{I}(\text{CF}_2)_4\text{I}$ ,  $\text{CH}_2\text{I}_2$ ,  $(\text{ICF}_2\text{CF}_2)_2\text{C}_6\text{F}_4$ ,  $\text{CF}_2\text{Br}_2$ ,  $\text{BrCH}_2\text{CH}_2\text{I}$ , and the like. It is preferable that those iodine- and/or bromine-containing monomers are used for the fluorine-containing polymers from the viewpoint of chain transfer reaction.

5        Particularly from the viewpoint of easiness of crosslinking reaction, iodine-containing monomers are preferable.

Examples of a polymerization method which can be used for those introducing methods are known methods such as emulsion polymerization, suspension polymerization and solution polymerization.

10        The polymers must contain iodine or bromine (a total amount thereof when the both are contained) in an amount of from 0.001 to 10 % by weight. When the amount is less than 0.001 % by weight, crosslinking cannot be carried out sufficiently, and when the amount is more than 10 % by weight, properties of the polymer such as heat  
15        resistance are lowered. The preferred content thereof is from 0.05 to 7 % by weight.

From the viewpoint of high crosslinking reactivity, it is preferable that the polymers mainly contain iodine rather than bromine.

In the present invention, the iodine- and/or bromine-  
20        containing polymers may be used alone or in a mixture of two or more thereof. The polymers may be blended to other polymer which does not contain iodine (bromine). Examples of the other non-iodine (non-bromine)-containing polymer which may be blended are, for instance, ultraviolet-crosslinkable polymers, such as epoxy acrylate, urethane  
25        acrylate, polyester acrylate and polyether acrylate.

The ultraviolet-crosslinkable polymer composition of the present invention comprises the above-mentioned iodine- and/or

bromine-containing polymer, a photoinitiator and a polyfunctional unsaturated compound.

The photoinitiator to be used in the present invention acts as a crosslinking agent, and photoinitiators which have been known in ultraviolet crosslinking can be used. Examples thereof are acetophenone initiators such as chloroacetophenone, diethoxyacetophenone,  $\alpha$ -aminoacetophenone, and hydroxyacetophenones, i.e. 2-hydroxy-2-methyl-1-phenylpropane-1-one, 1-(4-isopropylphenyl)-2-hydroxy-2-methylpropane-1-one and 1-hydroxycyclohexylphenylketone; benzoin initiators such as benzoin, benzoin ether and dimethylbenzyl ketal; benzophenone initiators such as benzophenone, benzoylbenzoic acid, hydroxybenzophenone, 4-phenylbenzophenone and acrylated benzophenone; thioxanthone initiators such as thioxanthone, 2-methylthioxanthone and 2,4-dimethylthioxanthone;  $\alpha$ -acyloxime ester, acylphosphine oxide, benzyl, camphorquinone, 2-ethylantraquinone, Michler's ketone, and the like.

An amount of the photoinitiator is usually from about 0.05 to about 10 parts by weight (hereinafter referred to as "part"), preferably from 1 to 5 parts based on 100 parts of the above-mentioned iodine- and/or bromine-containing polymer.

In case of benzophenone or thioxanthone initiators, a hydrogen donor may be added as a photoinitiating aid. Examples of the hydrogen donor are, for instance, aliphatic amines such as triethanolamine, methyldiethanolamine and triisopropanolamine; aromatic amines such as 4,4'-diethylaminophenone, 4-dimethylaminoethyl benzoate and 4-dimethylamino(n-butoxy)ethyl benzoate, and the like.



An amount of the hydrogen donor is usually from about 0.05 to about 10 parts, preferably from 0.5 to 5 parts based on 100 parts of the above-mentioned polymer.

The polyfunctional unsaturated compound to be used in the present invention acts as a crosslinking aid. The polyfunctional unsaturated compound is not limited and is effective as far as it has a reactivity against both of a polymer radical derived from iodine and/or bromine and generated by irradiation of ultraviolet rays and a radical derived from the photoinitiator.

Examples of the preferable polyfunctional unsaturated compound are, for instance, various diacrylates, trimethylolpropane triacrylate (TMPTA), trimethylolpropane trimethacrylate, triallyl isocyanurate (TAIC), triallyl cyanurate, triallyl trimellitate, pentaerythritol triacrylate, pentaerythritol tetraacrylate, dipentaerythritol hexaacrylate, N,N'-m-phenylenebismaleimide, dipropargyl terephthalate, diallyl phthalate, tetraallyl terephthalamide, triallyl phosphate, and the like. Among them, those having three or more functional groups are particularly preferable from the viewpoint of easiness of crosslinking of the polymer.

An amount of the polyfunctional unsaturated compound is from about 0.05 to about 10 parts, preferably from 0.5 to 5 parts based on 100 parts of the above-mentioned polymer.

Also as the case demands, a filler, processing aid, antioxidant, age resister, antiozonant, ultraviolet ray absorber, and the like may be blended.

Examples of the filler are metal oxides such as magnesium oxide, calcium oxide, titanium oxide, silicon oxide and aluminum oxide,

metal hydroxides such as magnesium hydroxide, aluminum hydroxide and calcium hydroxide, carbonates such as magnesium carbonate, aluminum carbonate, calcium carbonate and barium carbonate, silicates such as magnesium silicate, calcium silicate, sodium silicate and aluminum silicate, sulfates such as aluminum sulfate, calcium sulfate and barium sulfate, synthetic hydrotalcite, metal sulfides such as molybdenum disulfide, iron sulfide and copper sulfide, diatom earth, asbestos, lithopone (zinc sulfide/barium sulfate), graphite, carbon black, carbon fluoride, calcium fluoride, coke, fluorine-containing resin, and the like.

Examples of the processing aid are higher fatty acids such as stearic acid, oleic acid, palmitic acid and lauric acid; higher fatty acid salts such as sodium stearate and zinc stearate; higher fatty acid amides such as stearic acid amide and oleic amide; higher fatty acid esters such as ethyl oleate; higher fatty acid amines such as stearylamine and oleilamine; petroleum wax such as carnauba wax and ceresine wax; polyglycols such as ethylene glycol, glycerine and diethylene glycol; aliphatic hydrocarbons such as vaseline and paraffin; silicone oil; silicone polymer; low molecular weight polyethylene; phthalic acid esters; phosphoric acid esters; rosin; (halogenated) dialkylamine; (halogenated) dialkylsulfone; surfactant; and the like.

Examples of the antioxidant, age resister and antiozonant are phenol compounds such as 2,5-di-t-amylhydroquinone, amine-ketone compounds such as 2,2,4-trimethyl-1,2-dihydroquinone, aromatic binary amine compounds such as 4,4'-bis( $\alpha,\alpha$ -dimethylbenzyl)diphenylamine, and the like.

Examples of the ultraviolet ray absorber are benzophenone

compounds such as 2,4-dihydroxybenzophenone, amine compounds such as bis(2,2,6,6-tetramethyl-4-piperizyl)sebacate, benzotriazole compounds such as 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, and the like. An adding amount thereof differs depending on kind of additives and should be an amount not impairing a crosslinking reaction with ultraviolet rays.

The composition of the present invention can be prepared by known mixing methods. When the polymer is a resin, the mixing may be carried out at a temperature of not less than a melting point thereof by using an internal mixer, segment extruder, two-screw extruder, or the like. When the polymer is an elastomer and a molecular weight thereof is high, kneading may be carried out with an internal mixer such as an open roll or kneader mixer, and when the molecular weight thereof is low, mixing may be carried out with a planetary mixer, portable mixer, or the like. As far as the preparation is carried out in the room, there is no serious problem, but it is desirable that the mixing is carried out under the condition that no photo energy activating the photoinitiator is irradiated. Also the mixing may be carried out in a solution by using a good solvent.

In the present invention, suitable ultraviolet rays are those having a wavelength of from 420 to 150 nm. Also those having a wavelength range covering a maximum absorption wavelength of the above-mentioned photoinitiator can be used.

The composition of the present invention can be subjected to crosslinking with radiation and electron beams. The composition may be crosslinked with radiation or electron beams and as the case demands, in combination with ultraviolet rays. An irradiation time

varies depending on an intensity of ultraviolet rays, kind of photoinitiator, etc. When the intensity of the irradiation is about 165 mW/cm<sup>2</sup>, the irradiation time of from about ten seconds to about five minutes is enough. The irradiation can be carried out at room temperature.

The composition of the present invention is formed into a crosslinked article by crosslinking with ultraviolet rays. The crosslinked article can be used for various known applications. Particularly when a fluorine-containing polymer is used as a component polymer, the crosslinked article can be used for various parts in automotive industry, airplane industry and semiconductor industry by making the use of its excellent heat resistance, oil resistance, amine resistance, chemical resistance, etc. Further when a fluorine-containing elastomer is used, by utilizing its heat resistance, oil resistance and amine resistance, the crosslinked article can be used suitably for parts for oil sealing, for example, a hose, oil seal, etc. for engine oil of automotive, and also by utilizing its property of releasing less outgas, the crosslinked article can be used suitably for a gasket for a magnetic disc production apparatus, sealing materials for semiconductor production apparatuses and lines, etc.

Also the composition of the present invention can be used as a coating material. Concretely by dissolving or dispersing the composition of the present invention in a solvent such as water or organic solvent, a coating material which can be crosslinked with ultraviolet rays (further as the case demands, electron beams) is obtained.

The crosslinked article of the present invention can be

applied on the following molded articles, but the application is not limited to them.

Applications of molded articles:

Packing, O-ring, hose, other sealing material, diaphragm and valve having amine resistance, oil resistance, chemical resistance, steam resistance and weather resistance in transportation means such as automotive, ship and airplane; similar packing, O-ring, sealing material, diaphragm, valve, hose, roll and tube in chemical plant; similar packing, O-ring, hose, sealing material, belt, diaphragm, valve, roll and tube in foods plant and foods processing machine (including domestic appliances); similar packing, O-ring, hose, sealing material, diaphragm, valve and tube in nuclear plant; similar packing, O-ring, hose, sealing material, diaphragm, valve, roll, tube, mandrel, cable, flexible joint, belt, rubber plate and weatherstrip in general industrial parts; roll, roll blade and belt for PPC copying machine, etc.

More concretely there are the following applications.

(i) Automotive applications

① For sealing

Crank shaft seal

Gear box seal

Power piston packing

Cylinder liner seal

Valve stem seal

Front pump seal for automatic transmission

Rear axle pinion seal

Gasket for universal joint

Pinion seal for speed meter

Piston cup for foot brake

O-ring and oil seal for torque transmission

Seal for after-burner for exhaust gas

Bearing seal

5    ② For hose

EGR tube

Twin-carburetor tube

③ For diaphragm

Diaphragm for sensor of carburetor

10    ④ Other applications

Vibration proof rubber (engine mount, exhaust system, etc.)

Hose for after burner

(ii) Applications in chemical industries

① For sealing

15        Seals for pump, flow meter and pipe for chemicals

Seal for heat exchanger

Packing of glass cooler for sulfuric acid manufacturing  
equipment

20        Seals for sprinkler and transfer pump for agricultural  
chemicals

Seal for gas pipe

Seal for plating solution

Packing for high temperature vacuum dryer

Roll seal of belt for paper making

25        Seal for fuel battery

Joint seal for air duct

② For roll



Roll having trichlene resistance (for dyeing of fiber)

③ Other applications

Acid resistant hose (for concentrated sulfuric acid)

Packing for joint of tubes for gas chromatograph and pH  
meter

Chlorine gas transfer hose

Rainwater drain hoses for benzene and toluene reservoir  
tanks

Seal, tube, diaphragm and valve parts for analyzer and  
physical and chemical appliances

(iii) Applications in industrial machinery

① For sealing

Seals for hydraulic and lubricating machine

Bearing seal

Seal for dry copying machine

Seals for window, etc. of dry cleaner

Seal for equipment for concentrating uranium hexafluoride

Seal (vacuum) valve for cyclotron

Seal for automatic packaging machine

② Other applications

Rolls, scraper, tube and valve parts for printing equipment

Rolls, scraper, tube and valve parts for coating equipment

Ink tube, roll and belt for printer

Belt and rolls for dry copying machine

Diaphragms for pumps for analyzing sulfurous acid gas and  
chlorine gas in the air (environmental pollution-related  
meters)

Rolls and belt for printer

Squeeze rolls for pickling

(iv) Airplane applications

Valve stem seal for jet engine

5 Fuel feeding hose, gasket and O-ring

Rotating shaft seal

Gasket for hydraulic equipment

Seal for fire wall

(v) Ship applications

10 Stern seal for screw propeller shaft

Suction and exhaust valve stem seals for diesel engine

Valve seal for butterfly valve

Stem seal for butterfly valve

(vi) Food and medicine applications

15 Seal for plate heat exchanger

Solenoid valve seal for vending machine

Plugs for chemicals

(vii) Electrical applications

20 Insulation oil cap for a train of the Shikansen line

Venting seal for liquid ring transmission

Jacket for oil well cable

25 Further the molded article of the present invention can be used for O-ring, sealing material, hose, tube, diaphragm, roll, lining and coating in equipment for producing semiconductor, liquid crystal panel, plasma display panel, plasma address liquid crystal panel, field emission display panel, substrate for solar battery, etc., for example, CVD equipment, etching equipment, oxidation/diffusion equipment,



sputtering equipment, ashing equipment, ion implantation equipment, exhausting equipment, and the like which require plasma resistance; O-ring, sealing material, hose, tube, diaphragm and roll in wet etcher, cleaning equipment, pipes for chemicals, gas pipes, and the like which  
5 require chemical resistance; and further O-ring, sealing material, hose, tube, diaphragm and roll to be used on parts of the mentioned equipment which are required to be free from dust and metal.

Examples of other applications where chemical resistance is required are O-ring, sealing material, hose, tube, diaphragm of pump  
10 and wafer transferring rolls for resist developing solution, releasing solution and wafer cleaning solution in production equipment for semiconductor, liquid crystal panel, plasma display panel, etc.

When the polymer is a low molecular weight polymer, molding or processing can be carried out easily at working site by using  
15 an extrusion gun such as a hot melt gun or a dispenser. In that case, when the polymer is a fluorine-containing elastomer, it is preferable that a number average molecular weight thereof is within a range of from 1,000 to 50,000. When the number average molecular weight is less than 1,000, the crosslinking becomes difficult, and when the number  
20 average molecular weight is more than 50,000, the extrusion becomes difficult.

Examples of the application for work which can be carried out at site are coating of metal gasket for automotive engine, gasket for oil pan of engine, roll of copying machine or printer, sealing for building,  
25 sealing of gasket for magnetic recorder, sealing of filter unit of clean room, coating of printed circuit board, fixing of electric and electronic parts, insulation and water-proof treatment of lead wire terminal of

electrical equipment, sealing of an oven such as an electric oven,  
terminal treatment of sheath type heater, sealing of window frame of  
microwave oven, adhesion of CRT wedge and neck, adhesion of  
instrumentation parts of automotive, sealing of joints of kitchen, bath  
5 room and lavatory, etc.

The present invention is then explained concretely by means  
of examples, but is not limited thereto.

#### PREPARATION EXAMPLE 1

10 A 3-liter autoclave equipped with a thermometer, stirrer,  
nitrogen gas feeding tube and pressure reducing device was charged  
with 1,500 ml of pure water and 2 g of ammonium perfluorooctanoate.  
After the inside of the autoclave was sufficiently replaced with a  
vinylidene fluoride/hexafluoropropylene/tetrafluoroethylene gas  
15 mixture (hereinafter referred to as VdF/HFP/TFE, mole ratio: 18/71/11),  
a pressure and temperature of the inside of the autoclave were increased  
to 16 kg/cm<sup>2</sup>G and 80°C, respectively. Then 10 ml of a 0.3 % aqueous  
solution of ammonium persulfate was introduced under pressure with  
stirring. Since a pressure drop arose by the polymerization reaction,  
20 when the inside pressure decreased to 15 kg/cm<sup>2</sup>G, a chain transfer  
agent I(CF<sub>2</sub>)<sub>4</sub>I (1.3g) was introduced under pressure. At the time when  
the inside pressure was further decreased to 14 kg/cm<sup>2</sup>G, it was again  
increased to 16 kg/cm<sup>2</sup>G with a VdF/HFP/TFE gas mixture (50/20/30  
in mole ratio). Thereafter the polymerization was continued while  
25 adjusting the inside pressure within the range of from 14 to 16 kg/cm<sup>2</sup>G  
in such a manner.

When a total pressure drop reached 5 kg/cm<sup>2</sup>G after starting

of the polymerization reaction, 1.8 g of  $\text{CF}_2=\text{CFOCF}_2\text{CF}_2\text{CH}_2\text{I}$  was introduced. The polymerization was continued within the pressure range of from 14 to 16  $\text{kg/cm}^2\text{G}$  similarly. When a reduction of an amount of the gas mixture in a bomb reached 400 g, the inside pressure was liberated to terminate the polymerization reaction. The obtained dispersion was frozen, coagulated and dried to obtain a polymer (hereinafter referred to as "Polymer A").

As a result of NMR analysis, components of the obtained polymer was  $\text{VdF/HFP/TFE}=50/20/30$  % by mole. According to GPC measurement, a number average molecular weight thereof was 140,000 (based on polystyrene). The iodine content thereof was 0.55 % by weight according to an elementary analysis.

#### PREPARATION EXAMPLE 2

A 3-liter autoclave equipped with a thermometer, stirrer, nitrogen gas feeding tube and pressure reducing device was charged with 1,500 ml of pure water and 1.5 g of ammonium perfluorooctanoate. After the inside of the autoclave was sufficiently replaced with a vinylidene fluoride/hexafluoropropylene gas mixture (hereinafter referred to as  $\text{VdF/HFP}$ ) (mole ratio: 45/55), a pressure and temperature of the inside of the autoclave were increased to 14  $\text{kg/cm}^2\text{G}$  and 80°C, respectively. Then 4.5 g of  $\text{I}(\text{CF}_2\text{CF}_2)_2\text{I}$  as a polymerization initiator (25°C) was added and 10 ml of a 3 % aqueous solution of ammonium persulfate was introduced under pressure with stirring. Since a pressure drop arose by the polymerization reaction, when the inside pressure decreased to 13  $\text{kg/cm}^2\text{G}$ , the inside pressure was again increased to 15  $\text{kg/cm}^2\text{G}$  with a  $\text{VdF/HFP}$  gas mixture (78/22 in mole

ratio). Thereafter the polymerization was continued while adjusting the inside pressure within the range of from 13 to 15 kg/cm<sup>2</sup>G in such a manner. When a reduction of an amount of the gas mixture in a bomb reached 400 g, the inside pressure was liberated to terminate the polymerization reaction. The obtained dispersion was frozen, coagulated and dried to obtain a polymer (hereinafter referred to as "Polymer B").

As a result of NMR analysis, components of the obtained polymer was VdF/HFP=78/22 % by mole. According to GPC measurement, a number average molecular weight thereof was 41,000 (based on polystyrene). The iodine content thereof was 0.6 % by weight according to an elementary analysis.

### PREPARATION EXAMPLE 3

A 1-liter separable flask equipped with a thermometer, stirrer, nitrogen gas feeding tube and pressure reducing device was charged with 400 g of pure water, 0.2 g of sodium bicarbonate, 1 g of sodium dodecylsulfate, 1 g of polyoxyethylene nonyl phenyl ether and 100 g of a monomer mixture (ethyl acrylate/2-iodoethyl acrylate = 99/1 % by weight). After the inside of the flask was sufficiently replaced with nitrogen gas, 0.01 g of sodium hydrosulfite, 0.002 g of sodium formaldehyde sulfoxylate and 0.005 g of t-butyl hydroperoxide were added to initiate a polymerization reaction at 35°C. After six-hour reaction had been carried out so that a conversion of the polymerization became not less than 99 %, the reaction product was subjected to salting-out, washing with water and drying to obtain a polymer (hereinafter referred to as "Polymer C").

The iodine content of the obtained polymer was 0.43 % by weight as a result of elementary analysis.

#### PREPARATION EXAMPLE 4

5 A 1-liter separable flask equipped with a thermometer, stirrer, nitrogen gas feeding tube and pressure reducing device was charged with 400 g of pure water, 0.2 g of sodium bicarbonate, 1 g of sodium dodecylsulfate, 1 g of polyoxyethylene nonyl phenyl ether and 100 g of a monomer (ethyl acrylate = 100 % by weight). After the inside of the  
10 flask was sufficiently replaced with nitrogen gas, 0.01 g of sodium hydrosulfite, 0.002 g of sodium formaldehyde sulfoxylate and 0.005 g of t-butyl hydroperoxide were added to initiate a polymerization reaction at 35°C. After six-hour reaction had been carried out so that a conversion of the polymerization became not less than 99 %, the reaction product  
15 was subjected to salting-out, washing with water and drying to obtain a polymer (hereinafter referred to as "Polymer D").

#### EXAMPLE 1

To 100 parts of Polymer A prepared in Preparation Example 1  
20 were mixed 1.4 parts of 1-hydroxycyclohexyl phenylketone as a photoinitiator and 4.7 parts of trimethylolpropane triacrylate (TMPTA) as a polyfunctional unsaturated compound (crosslinking aid), and the mixture was kneaded and uniformly dispersed with an internal mixer. Then the obtained composition was subjected to pre-molding by  
25 pressing at a specific temperature to obtain an about 0.5 mm thick sheet.

The sheet was passed through a conveyor type ultraviolet ray

irradiation equipment to be subjected to irradiation with ultraviolet rays. Thereby a crosslinked article was obtained. With respect to the irradiation conditions, an intensity of ultraviolet rays was about 165 mW/cm<sup>2</sup>, an irradiation time was about 30 seconds and an ambient temperature was room temperature (about 25°C).

With respect to the crosslinked article, solubility in acetone (the crosslinked article was dipped in acetone at room temperature for three hours with stirring, and solubility was observed with naked eyes) and physical properties in normal state according to JIS K-6301 were measured. The results are shown in Table 1.

#### EXAMPLES 2 to 5

Components shown in Table 1 were mixed and pre-molded in the same manner as in Example 1 to obtain an about 0.5 mm thick sheet. The sheet was subjected to ultraviolet ray irradiation under the same irradiation conditions as in Example 1 to obtain a crosslinked article and its solubility in acetone and physical properties in normal state were determined. The results are shown in Table 1.



TABLE 1

| Polymer composition<br>(part by weight) | Example |     |     |      |     |
|---|---------|-----|-----|------|-----|
|   | 1       | 2   | 3   | 4    | 5   |
| Polymer                                 |         |     |     |      |     |
| Polymer A                               | 100     | 100 | 100 | 100  | 100 |
| Polymer B                               |         |     |     |      |     |
| Polymer C                               |         |     |     |      |     |
| Photoinitiator                          |         |     |     |      |     |
| 1-hydroxycyclohexyl<br>phenylketone     | 1.4     | 4.2 |     |      | 4.2 |
| 2,2-dimethoxy-2-<br>phenylacetone       |         |     | 2.7 |      |     |
| Benzophenone                            |         |     |     | 1.9  |     |
| Photoinitiating aid                     |         |     |     |      |     |
| Triethanolamine                         |         |     |     | 0.63 |     |
| Polyfunctional<br>unsaturated compound  |         |     |     |      |     |
| TMPTA                                   | 4.7     |     | 4.7 | 4.7  | 4.7 |
| TAIC                                    |         | 4   |     |      |     |

- continued -

- continued -

| Physical properties of<br>crosslinked article |  |           |           |           |           |
|---|--|-----------|-----------|-----------|-----------|
| Solubility in acetone                         |  | Insoluble | Insoluble | Insoluble | Insoluble |
| Physical properties in<br>normal state        |  |           |           |           |           |
| 100 % tensile stress<br>(MPa)                 |  | 3         | 1         | 1         | 1         |
| Strength at break (MPa)                       |  | 6         | 4         | 5         | 4.5       |
| Elongation at break (%)                       |  | 700       | 540       | 520       | 550       |
| Hardness (JIS A)                              |  | 56        | 55        | 55        | 54        |



### EXAMPLES 6 to 8

Components shown in Table 2 were mixed and pre-molded in the same manner as in Example 1 to obtain an about 0.5 mm thick sheet. In Examples 6 and 7, Polymer B prepared in Preparation Example 2 was  
5 used as an iodine-containing polymer, and in Example 8, Polymer C prepared in Preparation Example 3 was used as an iodine-containing polymer.

The sheet was subjected to ultraviolet ray irradiation under the same irradiation conditions as in Example 1 to give a crosslinked  
10 article and its solubility in acetone and physical properties in normal state were determined. The results are shown in Table 2.

TABLE 2

| Polymer composition<br>(part by weight) | Example |     |     |
|---|---------|-----|-----|
|   | 6       | 7   | 8   |
| Polymer                                 |         |     |     |
| Polymer A                               |         |     |     |
| Polymer B                               | 100     | 100 |     |
| Polymer C                               |         |     | 100 |
| Photoinitiator                          |         |     |     |
| 1-hydroxycyclohexyl<br>phenylketone     | 4.2     | 4.2 | 2.1 |
| 2,2-dimethoxy-2-<br>phenylacetone       |         |     |     |
| Benzophenone                            |         |     |     |
| Photoinitiating aid                     |         |     |     |
| Triethanolamine                         |         |     |     |
| Polyfunctional unsaturated<br>compound  |         |     |     |
| TMPTA                                   |         |     |     |
| TAIC                                    | 4       | 8   | 4   |

- continued

- continued -

| Physical properties of crosslinked article |  |           |           |
|--|--|-----------|-----------|
| Solubility in acetone                      |  | Insoluble | Insoluble |
| Physical properties in normal state        |  |           |           |
| 100 % tensile stress (MPa)                 |  | 0.3       | 0.5       |
| Strength at break (MPa)                    |  | 2         | 5         |
| Elongation at break (%)                    |  | 900       | 370       |
| Hardness (JIS A)                           |  | 36        | 50        |
|  |  |           | 58        |

### COMPARATIVE EXAMPLES 1 to 5

Components shown in Table 3 were mixed and pre-molded in the same manner as in Example 1 to obtain an about 0.5 mm thick sheet.

In Comparative Examples 1 and 2, a fluorine-containing elastomer of

5 VDF/HFP/TFE terpolymer (DAIEL G501 available from DAIKIN INDUSTRIES, LTD.) containing no iodine was used as a polymer, in

Comparative Example 3, Polymer A prepared in Preparation Example 1 was used as a polymer (no photoinitiator was used), in Comparative

Example 4, Polymer A was used as a polymer (no polyfunctional

10 unsaturated compound was used), and in Comparative Example 5,

Polymer D prepared in Preparation Example 4 and having neither iodine nor bromine was used as a polymer.

The sheet was subjected to ultraviolet ray irradiation under the same irradiation conditions as in Example 1 to obtain a crosslinked

15 article and its solubility in acetone and physical properties in normal state were determined. The results are shown in Table 3.

TABLE 3

| Polymer composition<br>(part by weight) | Comparative Example |     |     |     |     |
|---|---------------------|-----|-----|-----|-----|
|   | 1                   | 2   | 3   | 4   | 5   |
| Polymer                                 |                     |     |     |     |     |
| G501                                    | 100                 | 100 |     |     |     |
| Polymer A                               |                     |     | 100 | 100 |     |
| Polymer D                               |                     |     |     |     | 100 |
| Photoinitiator                          |                     |     |     |     |     |
| 1-hydroxycyclohexyl<br>phenylketone     | 2.1                 | 2.1 |     | 2.1 | 2.1 |
| 2,2-dimethoxy-2-<br>phenylacetone       |                     |     |     |     |     |
| Benzophenone                            |                     |     |     |     |     |
| Photoinitiating aid                     |                     |     |     |     |     |
| Triethanolamine                         |                     |     |     |     |     |
| Polyfunctional unsaturated<br>compound  |                     |     |     |     |     |
| TMPTA                                   | 4.7                 |     |     |     |     |
| TAIC                                    |                     | 4   | 4   |     | 4   |

- continued -

- continued -

Physical properties of  
crosslinked article

Solubility in acetone  
Physical properties in  
normal state

Dissolved Dissolved Dissolved Dissolved Dissolved

100 % tensile stress (MPa)

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Strength at break (MPa)

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Plastic  
deformation

Elongation at break (%)

Unmeasurable

Unmeasurable

Unmeasurable

Unmeasurable

Unmeasurable

Hardness (JIS A)

Unmeasurable

Unmeasurable

Unmeasurable

Unmeasurable

Unmeasurable

### INDUSTRIAL APPLICABILITY

According to the present invention, by using iodine and/or bromine as a crosslinking point, it is possible to carry out crosslinking of various polymers including fluorine-containing polymers which have  
5 been very difficult to crosslink with ultraviolet rays, and molding or processing can be made easily at working site at low temperature.

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## CLAIMS

1. A polymer composition which is crosslinkable with ultraviolet rays and comprises a polymer containing 0.001 to 10 % by weight of iodine and/or bromine, a photoinitiator as a crosslinking agent and a polyfunctional unsaturated compound as a crosslinking aid.

2. The polymer composition of Claim 1, wherein the polymer is a fluorine-containing polymer.

3. The polymer composition of Claim 2, wherein the fluorine-containing polymer is a fluorine-containing elastomer.

4. A crosslinked article obtained by subjecting the ultraviolet-crosslinkable polymer composition of any of Claims 1 to 3 to ultraviolet irradiation.

5. A gasket for magnetic disc production apparatus which is obtained by subjecting the ultraviolet-crosslinkable polymer composition of any of Claims 1 to 3 to ultraviolet irradiation.

6. A fluorine-containing polymer coating material comprising the ultraviolet-crosslinkable polymer composition of Claim 2 or 3.



# ABSTRACT

An ultraviolet-crosslinkable polymer composition comprising a polymer containing iodine and/or bromine in an amount of from 0.001 to 10 % by weight, a photoinitiator as a crosslinking agent and a polyfunctional unsaturated compound as a crosslinking aid. By using iodine and/or bromine as a crosslinking point, it is possible to carry out crosslinking of various polymers including fluorine-containing polymers which have been very difficult to crosslink with ultraviolet rays, and molding or processing can be made easily at working site at low temperature.

## PATENT COOPERATION TREATY

PCT

NOTIFICATION OF THE RECORDING  
OF A CHANGE(PCT Rule 92bis.1 and  
Administrative Instructions, Section 422)

From the INTERNATIONAL BUREAU

To:

ASAHINA, Sohta  
NS Building  
2-22, Tanimachi 2-chome  
Chuo-ku, Osaka-shi  
Osaka 540-0012  
JAPON

|  |   |
|--|---|
| Date of mailing (day/month/year)<br>11 février 2002 (11.02.02) | <b>IMPORTANT NOTIFICATION</b>   |
| Applicant's or agent's file reference<br>FP-6695PCT            |   |
| International application No.<br>PCT/JP00/05780                | International filing date (day/month/year)<br>28 août 2000 (28.08.00) |

## 1. The following indications appeared on record concerning:

☒ the applicant ☒ the inventor ☐ the agent ☐ the common representative

|  |                            |                          |
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## 2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☐ the person ☐ the name ☒ the address ☐ the nationality ☐ the residence

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## 3. Further observations, if necessary:

## 4. A copy of this notification has been sent to:

☒ the receiving Office ☐ the designated Offices concerned  
☐ the International Searching Authority ☒ the elected Offices concerned  
☐ the International Preliminary Examining Authority ☐ other:

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| The International Bureau of WIPO<br>34, chemin des Colombettes<br>1211 Geneva 20, Switzerland | Authorized officer<br><br>Shinji IGARASHI |
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**DECLARATION AND POWER OF ATTORNEY FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63)**

As a below named inventor, I hereby declare that: My residence, mailing address, and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ULTRAVIOLET-CROSSLINKABLE POLYMER COMPOSITION

the application of which

☐ is attached hereto

OR

☒ was filed on August 28, 2000 as United States Application Number or PCT International Application Number PCT/JP00/05780 (Confirmation No. \_\_\_\_\_), and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified application, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56, including for continuation-in-part application(s), material information which became available between the filing date of the prior application and the national or PCT international filing date of the continuation-in-part application.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or (f), or 365(b) of any foreign application(s) for patent, inventor's or plant breeder's rights certificate(s), or 365(a) of any PCT international application(s) which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application(s) for patent, inventor's or plant breeder's rights certificate(s), or any PCT international application(s) having a filing date before that of the application on which priority is claimed.

| Prior Foreign Application Number(s) | Country      | Foreign Filing Date    | Priority Claimed                    |                          |
|-------------------------------------|--------------|------------------------|-------------------------------------|--------------------------|
|                                     |              |                        | Yes                                 | No                       |
| <u>244653/1999</u>                  | <u>Japan</u> | <u>August 31, 1999</u> | <input checked="" type="checkbox"/> | <input type="checkbox"/> |

I hereby claim domestic priority benefits under 35 United States Code §120 of any United States application(s), §119(e) of any United States provisional application(s), or §365(c) of any PCT International application(s) designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in a listed prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge my duty to disclose any information material to the patentability of this application as defined in 37 C.F.R. 1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

| Prior U.S. or International Application Number(s) | U.S. or International Filing Date | Status |
|---|-----------------------------------|--------|
| _____   | _____                             | _____  |

I hereby appoint all attorneys of **SUGHRUE MION, PLLC** who are listed under the USPTO Customer Number shown below as my attorneys to prosecute this application and to transact all business in the United States Patent and Trademark Office connected therewith, recognizing that the specific attorneys listed under that Customer Number may be changed from time to time at the sole discretion of Sughrue Mion, PLLC, and request that all correspondence about the application be addressed to the address filed under the same USPTO Customer Number.



**23373**

PATENT TRADEMARK OFFICE

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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|  |       |                        |             |
| <b>NAME OF FIFTH INVENTOR:</b>   |       |                        |             |
| Given Name<br>(first and middle [if any])  |       | Family Name or Surname |             |
|  |       |                        |             |
| Inventor's Signature   |       | Date                   |             |
|  |       |                        |             |
| Residence: City  | State | Country                | Citizenship |
|  |       |                        |             |
| Mailing Address:   |       |                        |             |
| City   | State | Zip                    | Country     |
|  |       |                        |             |